

A theoretical determination of the diffusion-like ionisation time of Rydberg atoms

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Abstract. The non-stationary multistep (diffusion-like) ionisation of Rydberg atoms is considered theoretically. The investigation is based on the time-dependent Fokker–Planck equation and is related to collisional and microwave ionisation of Rydberg atoms. Analytical expressions for the mean time and the higher moments of the distribution of the diffusive ionisation times are derived. The conditions for the occurrence of diffusive ionisation are obtained. The relation of the present study to stationary diffusive ionisation and the experimental investigation of the ionisation of Rydberg atoms is discussed.

1. Introduction

Considerable attention is devoted at present to the multistep transitions between the Rydberg states of atoms (see e.g. Biberman *et al* 1982, Delone *et al* 1978, 1983, Kaulakys *et al* 1984, Jensen 1984, Bayfield and Pinnaduwege 1985a, b, van Leeuwen *et al* 1985). In such a process the change of the energy of a Rydberg atom in each elementary event (the absorption or stimulated emission of a low-frequency photon or the collision of a Rydberg atom with an atomic particle) is small in comparison with the binding energy of the Rydberg electron. The ionisation of the Rydberg atom and the recombination of the electron occur as a result of a great number of elementary events and are described by the Fokker–Planck equation (see Pitayevsky 1962, Lifshitz and Pitayevsky 1979, Biberman *et al* 1982, Kaulakys *et al* 1984). Until now a great deal of attention has been devoted to the theoretical investigation of the kinetics of Rydberg state occupation numbers, the ionisation rates of Rydberg atoms and the recombination of electrons in slightly ionised plasmas in stationary conditions only. Under the impulsive excitation of an atom to a high Rydberg state, as is the case in the experiments on the microwave ionisation of highly excited hydrogen atoms (Bayfield and Pinnaduwege 1985a, b, van Leeuwen *et al* 1985), the rate of diffusive ionisation of the Rydberg atom is defined by the solution of the non-stationary Fokker–Planck equation with specified initial and boundary conditions. In general, the non-stationary Fokker–Planck equation, in the case when the diffusion and drift coefficients are dependent on the energy, cannot be solved analytically, and one must be satisfied with the estimation of the order of the mean time of the diffusive ionisation only (Delone *et al* 1978, 1983).

The purpose of the present work is to obtain analytical expressions for the mean time and higher moments of the distribution of the diffusive ionisation times of the Rydberg atoms. In addition, the conditions for the occurrence of diffusive ionisation will be obtained and the relation between stationary diffusive ionisation and the

experimental investigation of the collisional and microwave ionisation of the Rydberg atoms will be discussed.

2. Diffusive approximation

In the situation when either the change in energy $\Delta\varepsilon$ of a Rydberg atom in one collision event with an atomic particle or when the absorption or stimulated emission of the low-frequency photon is small in comparison with the binding energy of the Rydberg electron $\varepsilon = \frac{1}{2}n^{-2}$, the process of the multistep transitions between the Rydberg states of atoms is diffusion-like. The theoretical treatment of the evolution of the probability density $f(\varepsilon, t)$ of the electrons in the energy space of the Rydberg atom leads to a Fokker-Planck-type diffusion equation (Lifshitz and Pitayevsky 1979)

$$\frac{\partial f(\varepsilon, t)}{\partial t} = \frac{\partial}{\partial \varepsilon} \left(\mathcal{D}(\varepsilon) \frac{\partial f(\varepsilon, t)}{\partial \varepsilon} \right) \quad (1)$$

where $f_0(\varepsilon)$ is the steady-state solution of equation (1), $\mathcal{D}(\varepsilon) = B(\varepsilon)f_0(\varepsilon)$, and $B(\varepsilon)$ is the diffusion coefficient of electrons in the energy space, given by

$$B(\varepsilon) = \frac{1}{2} \sum_{n'} K_{n,n'} (\Delta\varepsilon_{n,n'})^2. \quad (2)$$

Here $K_{n,n'}$ is the $n \rightarrow n'$ transition rate, and $\Delta\varepsilon_{n,n'} = \varepsilon' - \varepsilon$.

For the collisions of the Rydberg atoms with neutral atomic particles

$$K_{n,n'} = \langle v\sigma_{n,n'} \rangle N \quad (3)$$

where v is the relative collision velocity, N is the density of atomic particles, $\sigma_{n,n'}$ is the cross section for a $n \rightarrow n'$ transition and the average is taken over the distribution of the relative collision velocity.

A simple analytical expression for the cross section of the state-changing collisions of Rydberg atoms with neutral atomic particles is given by Kaulakys (1985, 1986) on the basis of the free-electron model

$$\sigma_{nl,n'} = \frac{\pi}{v^2 n'^3} \int_{p_i}^{\infty} |g_{nl}(p)|^2 p \, dp \int_{2p_i}^{2p} |f_e(p, Q)|^2 dQ \quad p_i = \frac{|\Delta\varepsilon_{nl,n'}|}{2v}. \quad (4)$$

Here $g_{nl}(p)$ is the radial wavefunction of the Rydberg electron in momentum space and $f_e(p, Q)$ is the electron-perturber scattering amplitude for a given momentum transfer Q . Substitution of equations (3) and (4) into equation (2) yields (see Kaulakys 1986)

$$B(\varepsilon) \approx \frac{2^{9/2} N \sigma_e v_T^{7/2} \xi_d^{3/2} (3 + 4\xi_d^2)}{\pi b^{3/2} (3 + 2\xi_d^2)^2} \quad (5)$$

where $b = 0.645$, σ_e is the electron-perturber elastic scattering cross section, $v_T = (2T/\mu)^{1/2}$ is the thermal velocity, and $\xi_d = b\varepsilon/v_T$. If $\xi_d \ll 1$, equation (5) reduces to the result of Pitayevsky (1962)

$$B_p(\varepsilon) = (2^{9/2}/3\pi) N \sigma_e v_T^2 \varepsilon^{3/2} \quad (6)$$

where $\sigma_e (= \sigma_e)$ is the momentum transfer cross section for electron-perturber elastic collisions.

The steady-state solution $f_0(\varepsilon)$ for such a process is the Boltzmann distribution (Lifshitz and Pitayevsky 1979, Kaulakys *et al* 1984):

$$f_0 = \varepsilon^{-5/2} \exp(\varepsilon/T). \quad (7)$$

Note that according to equation (2) the diffusion coefficient in the space of the principal quantum numbers $B_n = \frac{1}{2} \sum_{n'} K_{n,n'} (n' - n)^2$, when $|\Delta n| \ll n, n'$, is related to the diffusion coefficient in energy space by

$$B_n = B(\varepsilon) n^6 / Z^4 \quad \varepsilon = Z^2 / (2n^2) \quad (8)$$

where Z is the charge of the core of the Rydberg atom.

The latest experiments on the microwave ionisation of highly excited hydrogen atoms (Bayfield and Pinnaduwege 1985a, b, van Leeuwen *et al* 1985) indicate that the atom polarised along the direction of a microwave electric field responds to the field one dimensionally, and the multiphoton transitions between Rydberg states appear as a chaotic process. In such a case the steady-state solution is equal to the density of the states of the one-dimensional Rydberg atom

$$f_0 = \varepsilon^{-3/2} \quad (9)$$

and the diffusion coefficient for $\omega n^3 Z^{-2} \gg 1$ was given by Delone *et al* (1983) and Jensen (1984)

$$B_n = 0.27 F_0^2 n^3 / (\omega Z)^{4/3} \quad \omega n^3 Z^{-2} \gg 1. \quad (10)$$

Here F_0 and ω are the microwave electric field amplitude and angular frequency. According to equation (8) we have

$$B(\varepsilon) = 0.75 F_0^2 \varepsilon^{3/2} / (Z \omega^4)^{1/3}. \quad (11)$$

Similar investigation is possible for other processes involving the multistep transitions between the Rydberg states of atoms, such as Rydberg-electron inelastic collisions in a low-temperature plasma (see Biberman *et al* 1982) and, perhaps, for Rydberg-Rydberg and Rydberg-molecule inelastic collisions.

Under a non-stationary excitation of atoms to the high Rydberg states the rate of diffusive ionisation of the Rydberg atoms for such multistep processes is defined by the solution of the Fokker-Planck equation (1). In general, equation (1) with the diffusion coefficients and the function $f_0(\varepsilon)$ given by equations (5)-(11) (and so on) cannot be solved analytically. We shall, however, show in the next section that the mean time and the moments of the distribution of the diffusive ionisation times of Rydberg atoms may be obtained analytically.

3. Diffusive ionisation time

The Fokker-Planck equation (1) may be rewritten in the following form

$$\frac{\partial f(\varepsilon, t)}{\partial t} = -\frac{\partial}{\partial \varepsilon} (a(\varepsilon) f(\varepsilon, t)) + \frac{\partial^2}{\partial \varepsilon^2} (B(\varepsilon) f(\varepsilon, t)) \quad (12)$$

where the drift coefficient

$$a(\varepsilon) = B(\varepsilon) \frac{d}{d\varepsilon} \ln(B(\varepsilon) f_0(\varepsilon)) = \sum_{n'} K_{n,n'} \Delta \varepsilon_{n,n'}. \quad (13)$$

The distribution function $f(\varepsilon, t)$ is also the function of the initial electron energy ε_0

$$f(\varepsilon, t) = p(\varepsilon, t|\varepsilon_0, 0) \quad p(\varepsilon, 0|\varepsilon_0, 0) = \delta(\varepsilon - \varepsilon_0). \quad (14)$$

The function $p(\varepsilon, t|\varepsilon_0, 0)$, as a function of ε_0 , satisfies the backward Fokker-Planck equation (see e.g. Gardiner 1983)

$$\frac{\partial p(\varepsilon, t|\varepsilon_0, 0)}{\partial t} = a(\varepsilon_0) \frac{\partial p(\varepsilon, t|\varepsilon_0, 0)}{\partial \varepsilon_0} + B(\varepsilon_0) \frac{\partial^2 p(\varepsilon, t|\varepsilon_0, 0)}{\partial \varepsilon_0^2}. \quad (15)$$

The probability density of the time that the electron spends in the energy interval $[\varepsilon_1, \varepsilon_2]$, under the condition that at time $t=0$ the energy of the electron was ε_0 , is

$$G(\varepsilon_0, t) = \int_{\varepsilon_1}^{\varepsilon_2} p(\varepsilon, t|\varepsilon_0, 0) d\varepsilon \quad \varepsilon_1 \leq \varepsilon_0 \leq \varepsilon_2. \quad (16)$$

Integration of equation (15) over the energy ε according to equation (16) yields

$$\frac{\partial G(\varepsilon_0, t)}{\partial t} = a(\varepsilon_0) \frac{\partial G(\varepsilon_0, t)}{\partial \varepsilon_0} + B(\varepsilon_0) \frac{\partial^2 G(\varepsilon_0, t)}{\partial \varepsilon_0^2}. \quad (17)$$

The moment of order k for the distribution of the times spent by the electron in the interval $[\varepsilon_1, \varepsilon_2]$ is given, according to equation (16), by

$$T_k(\varepsilon_0) \equiv \overline{t^k(\varepsilon_0)} = - \int_0^\infty t^k \frac{\partial G(\varepsilon_0, t)}{\partial t} dt = k \int_0^\infty t^{k-1} G(\varepsilon_0, t) dt. \quad (18)$$

In equation (18) it has been taken into account that $G(\varepsilon_0, 0) = 1$ and $G(\varepsilon_0, \infty) = 0$. Multiplying equation (17) by t^{k-1} and integrating over the time interval $(0, \infty)$, one obtains an equation for the moment $T_k(\varepsilon_0)$:

$$B(\varepsilon_0) \frac{d^2 T_k(\varepsilon_0)}{d\varepsilon_0^2} + a(\varepsilon_0) \frac{dT_k(\varepsilon_0)}{d\varepsilon_0} + k T_{k-1}(\varepsilon_0) = 0 \quad k = 1, 2, \dots \quad T_0 = 1. \quad (19)$$

Equation (19) must be supplemented by the boundary conditions corresponding to the Rydberg atom's ionisation process, such as

$$T_k(0) = 0 \quad \left. \frac{dT_k(\varepsilon_0)}{d\varepsilon_0} \right|_{\varepsilon_0 = \varepsilon_2} = 0. \quad (20)$$

The first of the conditions (20) implies the absence of free electrons and, consequently, the absence of electrons with very small binding energy, i.e. $f(\varepsilon, t)|_{\varepsilon \rightarrow 0} = 0$ (see e.g. Gardiner 1983). The second condition implies the absence of a flow of electrons for certain $\varepsilon_2 > \varepsilon_0$ (as a consequence of the large intervals between neighbouring energy levels of the Rydberg atom). As will be shown below, the solution of equation (19) is not usually very sensitive to the ε_2 in the second boundary condition.

The solution of equation (19) with boundary conditions (20) is of the following form

$$T_k(\varepsilon_0) = k \int_0^{\varepsilon_0} \frac{dy}{\mathcal{D}(y)} \int_y^{\varepsilon_2} f_0(x) T_{k-1}(x) dx. \quad (21)$$

Expression (21) represents a recurrence relationship and enables one to obtain any moment of the distribution of the Rydberg atom's diffusive ionisation time. Having obtained some of the moments, the distribution function may then be constructed according to the maximum-entropy principle (see e.g. Kociszewski 1985).

Among the most interesting practical cases, the dependence of f_0 and B on the energy ϵ is a power-type dependence (see, for example, equations (6), (7), (9), (11) and also Biberman *et al* (1982), Kaulakys *et al* (1984))†

$$f_0 = C\epsilon^{-r} \quad B = A\epsilon^s \tag{22}$$

where C , A , r and s are constants. The substitution of equation (22) into equation (21) yields

$$T(\epsilon_0) \equiv T_1(\epsilon_0) = \frac{\epsilon_0^{2-s}}{(2-s)(r-1)A} \left[1 - \frac{2-s}{r-s+1} \left(\frac{\epsilon_0}{\epsilon_2} \right)^{r-1} \right] \quad r-s+1 > 0 \quad 2-s > 0 \tag{23}$$

$$T_2(\epsilon_0) = \frac{\epsilon_0^{2(2-s)}}{(2-s)^2(r-1)(r+s-3)A^2} \left[1 - \frac{4(r-1)(2-s)}{(r-s+1)^2} \left(\frac{\epsilon_0}{\epsilon_2} \right)^{r+s-3} + \frac{2(2-s)(r+s-3)}{(r-s+1)(r-2s+3)} \left(\frac{\epsilon_0}{\epsilon_2} \right)^{r-1} \right] \tag{24}$$

and so on.

It should be noted that all of the moments of the distribution of the Rydberg atom's diffusive ionisation time are finite if $r-s+1 > 0$ and $2-s > 0$. The mean ionisation time and the other moments of the time for diffusive ionisation become infinite if either of these conditions is violated. On the other hand, the analysis of the solutions of the stationary Fokker-Planck equation (see Kaulakys *et al* (1984) and § 4 below) shows that diffusive ionisation takes place if $r-s+1 > 0$ and it becomes impossible when $r-s+1 < 0$.

According to equation (23) the mean time of the diffusive ionisation depends weakly on ϵ_2 if $r \gg 1$ or $\epsilon_2 \gg \epsilon_0$. When $\epsilon_2 = \epsilon_0$ and $\epsilon_2 \rightarrow \infty$ the quantity $T(\epsilon_0)$ gives, respectively, the lower and the upper bounds for the mean ionisation time.

Equation (19) may also be solved with other boundary conditions, for example

$$T_k(0) = 0 \quad T_k(\epsilon_2) = 0. \tag{25}$$

The second of the conditions (25) implies the absence of electrons with binding energy $\epsilon \geq \epsilon_2$. This may be due to the quenching of the Rydberg atoms with small principal quantum numbers, caused by radiative relaxation, and so on.

The solution of equation (19) with boundary conditions (25) is

$$T_k^{(0)}(\epsilon_0) = k \left[\left(\int_0^{\epsilon_0} \frac{d\epsilon}{\mathcal{D}(\epsilon)} \right) \int_0^{\epsilon_2} \frac{d\epsilon}{\mathcal{D}(\epsilon)} \int_0^\epsilon f_0(\epsilon') T_{k-1}(\epsilon') d\epsilon' - \left(\int_{\epsilon_0}^{\epsilon_2} \frac{d\epsilon}{\mathcal{D}(\epsilon)} \right) \int_0^{\epsilon_0} \frac{d\epsilon}{\mathcal{D}(\epsilon)} \int_0^\epsilon f_0(\epsilon') T_{k-1}(\epsilon') d\epsilon' \right] \left(\int_0^{\epsilon_2} \frac{d\epsilon}{\mathcal{D}(\epsilon)} \right)^{-1}. \tag{26}$$

The substitution of equations (22) into equation (26) yields

$$T^{(0)}(\epsilon_0) = \frac{\epsilon_0^{2-s}}{(2-s)(r-1)A} \left[1 - \left(\frac{\epsilon_0}{\epsilon_2} \right)^{r-1} \right] \quad r-s+1 > 0 \quad 2-s > 0 \tag{27}$$

$$T_2^{(0)}(\epsilon_0) = \frac{(3+r-2s)\epsilon_0^{2(2-s)} + 2(r+s-3)\epsilon_2^{1-r}\epsilon_0^{3+r-2s} - 3(r-1)\epsilon_2^{3-r-s}\epsilon_0^{r-s+1}}{(2-s)^2(r-1)(r+s-3)(3+r-2s)A^2}. \tag{28}$$

† The analysis of formula (21) shows that the exponential in equation (7) may be neglected when $(\epsilon/T) \ll 1$.

Equations (26)–(28) do not represent the moments of the diffusive ionisation time but represent the moments for the distribution of the times that the electron spends in the interval $[0, \varepsilon_2]$ when the electron may escape from this interval through either end of the interval. In addition, the relative probability of the ionisation of the Rydberg atom is (Gardiner 1983, Kaulakys *et al* 1984)

$$P_i = \left(\int_{\varepsilon_0}^{\varepsilon_2} \frac{d\varepsilon}{\mathcal{D}(\varepsilon)} \right) \left(\int_0^{\varepsilon_2} \frac{d\varepsilon}{\mathcal{D}(\varepsilon)} \right)^{-1} \quad (29)$$

while the probability of the relaxation of the Rydberg atom to the ground state is

$$P_r = \left(\int_0^{\varepsilon_0} \frac{d\varepsilon}{\mathcal{D}(\varepsilon)} \right) \left(\int_0^{\varepsilon_2} \frac{d\varepsilon}{\mathcal{D}(\varepsilon)} \right)^{-1}. \quad (30)$$

The substitution of equations (22) into equations (29) and (30) gives

$$P_i = \begin{cases} 1 - (\varepsilon_0/\varepsilon_2)^{r-s+1} & r-s+1 > 0 \\ 0 & r-s+1 < 0 \end{cases} \quad (31)$$

$$P_r = 1 - P_i. \quad (32)$$

Thus, if $\varepsilon_2 \gg \varepsilon_0$ and $r-s+1 > 0$, the relative probability of ionisation is close to 1 and equations (26)–(28) reduce to equations (21), (23) and (24).

4. Discussion and relation with experiments

The theoretical cross sections for the direct ionisation of a Rydberg atom by a collision with a neutral atomic particle obtained on the bases of quasimolecular states (Duman and Shmatov 1980, Mihajlov and Janev 1981) and in the impulse approximation (Kaulakys 1985) are small for $n \sim 20$ –100. The efficient collisional ionisation of the alkali-metal Rydberg atoms with $n \sim 20$ –60 observed in experimental studies (Švedas and Stakišaitis 1984, Niemax 1983) cannot be explained on the bases of these theoretical approaches, and the diffusive mechanism for collisional ionisation of Rydberg atoms has been presented as an interpretation of the experimental results (Kaulakys *et al* 1984). However, the experiments on the collisional ionisation of the Rydberg atoms were performed under stationary conditions and the ionisation delay time was not measured. Therefore, in the paper by Kaulakys *et al* (1984) the stationary Fokker-Planck equation

$$\frac{\partial}{\partial \varepsilon} \left(\mathcal{D}(\varepsilon) \frac{\partial}{\partial \varepsilon} \frac{f(\varepsilon)}{f_0(\varepsilon)} \right) + \gamma \delta(\varepsilon - \varepsilon_0) - W(\varepsilon) = 0 \quad (33)$$

was solved, where $B(\varepsilon)$ and $f_0(\varepsilon)$ are given by equation (22) and the quenching rate of the Rydberg atoms is of the power-type form

$$W(\varepsilon) = f(\varepsilon) \varepsilon^q / \tau. \quad (34)$$

Here γ is the rate of generation of Rydberg atoms with energy ε_0 , and τ and q are constants. Equation (34) represents radiative relaxation of the Rydberg atoms if $q = \frac{3}{4}$ and quenching of the Rydberg atoms on the surfaces of the cell if $q = 0$. The efficiency

(probability) of the diffusive ionisation of the Rydberg atoms under stationary conditions is the ratio of the diffusive ionisation flow to the rate of generation of the Rydberg atoms

$$P_i = \frac{\mathcal{D}(\varepsilon)}{\gamma} \left. \frac{d f(\varepsilon)}{d\varepsilon f_0(\varepsilon)} \right|_{\varepsilon=0} \tag{35}$$

The solution of equation (33) with boundary conditions $f(0) = f(\varepsilon_2) = 0$ according to equations (22), (34) and (35) gives (see Kaulakys *et al* 1984)

$$P_i = \frac{y_0^\nu}{2^{\nu-1}\Gamma(\nu)} \left(K_\nu(y_0) - K_\nu(y_2) \frac{I_\nu(y_0)}{I_\nu(y_2)} \right) \tag{36}$$

where

$$y_i = \frac{2\varepsilon_i^{m/2}}{m(A\tau)^{1/2}} \quad i = 0, 2 \quad \nu = \frac{r-s+1}{m} > 0 \quad m = 2 - s + q > 0.$$

I_ν and K_ν are modified Bessel functions of the first and second kind, respectively, and Γ is the gamma function. If $\varepsilon_2 \gg \varepsilon_0$ equation (36) reduces to

$$P_i = K_\nu(y_0) y_0^\nu / (2^{\nu-1}\Gamma(\nu)) \quad \varepsilon_2 \gg \varepsilon_0 \tag{37}$$

and for $y_0 \ll 1$ we have

$$P_i = 1 - \frac{e_0^{2-s+q}}{(r-1-q)(2-s+q)A\tau} \quad 1 - P_i \ll 1. \tag{38}$$

Equations (36)–(38) allow one to explain qualitatively the experimental results of the stationary collisional ionisation of the Rydberg atoms (see Kaulakys *et al* 1984) while, for a quantitative interpretation, it is necessary to know the rate of quenching of the Rydberg atoms.

On the other hand, it may be shown that the quantity $T^{-1}(\varepsilon_0)$ characterises the mean rate of diffusive ionisation of the Rydberg atom. When the quenching rate of the Rydberg atoms does not depend on the energy ($q = 0$ in equation (34)) the efficiency of the diffusive ionisation may be evaluated as

$$P_i \approx T^{-1}(\varepsilon_0) / (T^{-1}(\varepsilon_0) + \tau^{-1}) \approx 1 - T(\varepsilon_0) / \tau \quad T(\varepsilon_0) \ll \tau. \tag{39}$$

The substitution of equation (23) when $\varepsilon_2 \gg \varepsilon_0$ into equation (39) gives equation (38). This shows that the quantity $T^{-1}(\varepsilon_0)$ is indeed an appropriate rate of diffusive ionisation.

Under the impulsive excitation of an atom to a Rydberg state, the flow of diffusive ionisation depends on the time and the quantity $T^{-1}(\varepsilon_0)$ characterises the rate of diffusive ionisation of the Rydberg atom at time $t \sim T(\varepsilon_0)$ after the excitation. Note that the mean-square deviation or the variance $\sigma^2 = T_2 - T^2$ is a measure of the scatter of the distribution of diffusive ionisation times.

It should be noted, however, that the direct experimental proof of the diffusive collisional ionisation of Rydberg atoms is lacking and time-resolved experimental measurements of the collisional ionisation of the Rydberg atoms in gas are desirable. The experimental distribution of the ionisation times may be analysed on the basis of the present theory. In particular, if diffusive ionisation takes place, the mean delay time of the ionisation of the Rydberg atoms after the impulsive excitation to a Rydberg state with energy ε_0 must be equal to $T(\varepsilon_0)$ (obtained from equations (6), (7) and (23)):

$$T(\varepsilon_0) = \frac{1}{4}\pi\varepsilon_0^{1/2} / (2^{1/2}N\sigma_i v_T^2) \quad \varepsilon_0 \ll v_T. \tag{40}$$

An analogous situation is found in the field of experiments on the microwave ionisation of highly excited hydrogen atoms. The experimental studies performed until now were devoted either to the measurement of the field strengths at which the ionisation of the atoms appears or to the investigation of the dependence of ionisation on the frequency of the microwave field. As noted in the review paper by Delone *et al* (1983), there is no direct experimental evidence of the microwave diffusive ionisation of the Rydberg atoms and a time-resolved experiment is desirable.

The mean time of the diffusive ionisation given according to equations (9), (10) and (23)

$$T(\epsilon_0) = 5.3 F_0^{-2} (\omega^4 Z)^{1/3} \epsilon_0^{1/2} \quad \omega n^3 Z^{-2} \geq 1 \quad (41)$$

is in agreement with the experimental data (van Leeuwen *et al* 1985). Nevertheless, measurement of the time dependence of the microwave ionisation of the hydrogen Rydberg atoms is much needed if the theoretical model of the ionisation process is to be perfected.

To summarise, the time-dependent diffusion-like ionisation of the Rydberg atoms has been investigated theoretically. Analytical expressions for the mean time and higher moments of the distribution of diffusive ionisation times of the Rydberg atoms have been obtained and the conditions necessary for the occurrence of diffusive ionisation have been established. The analysis of the time-dependent ionisation process presented here establishes a theoretical basis for the direct experimental confirmation and investigation of the diffusion-like mechanism of the ionisation of Rydberg atoms.

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